

Science-based cleanup of Rocky Flats

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The chemical and physical interactions of radioactive compounds are key to understanding how they can contaminate the environment and, more importantly, how best to remove them.

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From 1952 to 1989, the Rocky Flats Nuclear Weapons Plant, located about 24 km northwest of Denver, Colorado, made components for the nation's nuclear arsenal using various radioactive materials, including plutonium and uranium; toxic metals such as beryllium; and hazardous solvents, degreasers, and other chemicals. The key component produced at Rocky Flats was the plutonium pit, commonly referred to as the trigger for a nuclear weapon. The pit provides energy to fuel the explosion.

In 1989 the Federal Bureau of Investigation and the Environmental Protection Agency abruptly halted nuclear production work to investigate environmental and safety concerns, and the site was added to the EPA's Superfund list later that year. In 1993 the secretary of energy announced the end of the nuclear production mission, and the area became known as the Rocky Flats Environmental Technology Site (RFETS) in 1995. Nearly 40 years of nuclear weapons production left behind contaminated facilities, soils, and surface and ground water.

More than 2.5 million people live within an 80-km radius of the site, and 300 000 of those live in the Rocky Flats watershed. The sudden shutdown left large quantities of plutonium and other hazardous substances in various stages of processing and storage. Because plutonium is dangerous to human health, even in minute quantities, the cleanup of plutonium-contaminated materials is highly complex, tedious, and labor intensive.

In March 1995 the US Department of Energy estimated that the cleanup for Rocky Flats would cost in excess of \$37 billion and take 70 years to complete. By 1996, DOE and independent contractor Kaiser-Hill Co had initiated a massive effort that eventually resulted in a credible plan to accelerate the closure of Rocky Flats by 31 December 2006 at a contracted cost of \$7 billion. After a troubled start, Kaiser-Hill completed the task nearly a year ahead of schedule.

What led to the turnaround? Without question, an incentive-laden contract, strong support and stable funding from Congress, high-level DOE support that mobilized the entire DOE complex to assist in the cleanup, technological innovation, and improved scientific understanding all contributed. Much has been made of the contractor's fee of more than \$500 million, but less has been said about the role that scientific understanding played in guiding key cleanup decisions and facilitating good project management.

The impetus to understand the science behind plutonium contamination gained momentum in 1995 when in-

tense rainfall and wet springtime conditions raised concerns about the mobility and dispersal of plutonium and americium. To account for increased concentrations of plutonium at various surface water-monitoring locations, researchers hypothesized that plutonium was soluble in surface and ground water. But modeling efforts at the time predicted very little movement of plutonium. The discord between the data and predictions prompted DOE and Kaiser-Hill in 1995 to establish the Actinide Migration Evaluation (AME) advisory group. The idea was to solicit advice and technical expertise on how elements such as plutonium, uranium, and americium are likely to behave in the air, surface water, ground water, and soil (see box 1).

Supported by scientific measurements, the group found that plutonium and americium form insoluble oxides and colloids that adhere to small organic and mineral particles in soil. The particles can migrate throughout the Rocky Flats environment by wind and surface water; particles are lifted from some location, suspended in air or water, and then re-deposited as sediment somewhere else. This understanding showed that soluble transport models were, in fact, not appropriate to describe the transport of plutonium and americium and led to the adoption of erosion and sediment-transport models. And it provided the basis for how best to negotiate a cleanup agreement and settle on an allowable standard of 50 picocuries per gram of soil. The relevant measure of plutonium and americium concentration is how much radiation is given off per unit volume or mass.

Did it save a lot of taxpayer dollars? That's difficult to determine. What everyone agrees on is that scientific understanding provided clarity and focus on the real issues surrounding plutonium and americium in the RFETS environment. The clarity and focus in turn allowed for good project management, guided remediation efforts, and most certainly helped shave decades and billions of dollars off the initial cleanup estimate.

Site details

Nearly the size of a small city with its own fire department, medical offices, cafeteria, and water- and sewage-treatment plants, Rocky Flats comprised more than 800 structures on a 1.6-square-kilometer industrial area surrounded by approximately 24 square kilometers of controlled open space (see figure 1). The open space continues to serve as a buffer between Rocky Flats and the nearby, growing communities and is home to many species of animals and plants.



Figure 1. A 1995 photograph of the Rocky Flats Environmental Technology Site shows how industrial the region had become as a nuclear production facility, with more than 800 structures built on 1.6 square kilometers, surrounded by 24 square kilometers of open space. The inset shows the same area in October 2005, after remediation.

Water at Rocky Flats and the surrounding area is distributed among surface water, shallow ground water, and deep ground water.¹ A series of detention ponds had been constructed along creeks to manage plant waste and surface water runoff. Shallow ground water refers to water within the alluvium and weathered bedrock and is found to a depth of 30 m. Water from the surface filters downward, recharging the shallow ground water, which in turn recharges the stream channels

at certain times of the year. Beneath the alluvium is highly impermeable bedrock that inhibits vertical flow. As a result, shallow ground water flows laterally and either discharges into the streams or emerges as hillside springs and seeps. Deep regional ground water flows about 200–300 m below the surface. Because of the intervening bedrock, that regional ground water aquifer is hydrologically isolated from the Rocky Flats surface and shallow ground water and from actinide contaminants.²

Box 1. Actinide migration evaluation

In 1995, to address the question of how actinide elements migrate in the Rocky Flats environment, Kaiser-Hill Co and the US Department of Energy commissioned the Actinide Migration Evaluation. Initially, AME advisers were recruited to evaluate and provide guidance on environmental conditions—in particular, the geochemistry of actinides at Rocky Flats, the way the radioactive elements there move from one location to another, and the role of erosion. The charter was rapidly expanded to include recommendations for long-term protection of surface-water quality.

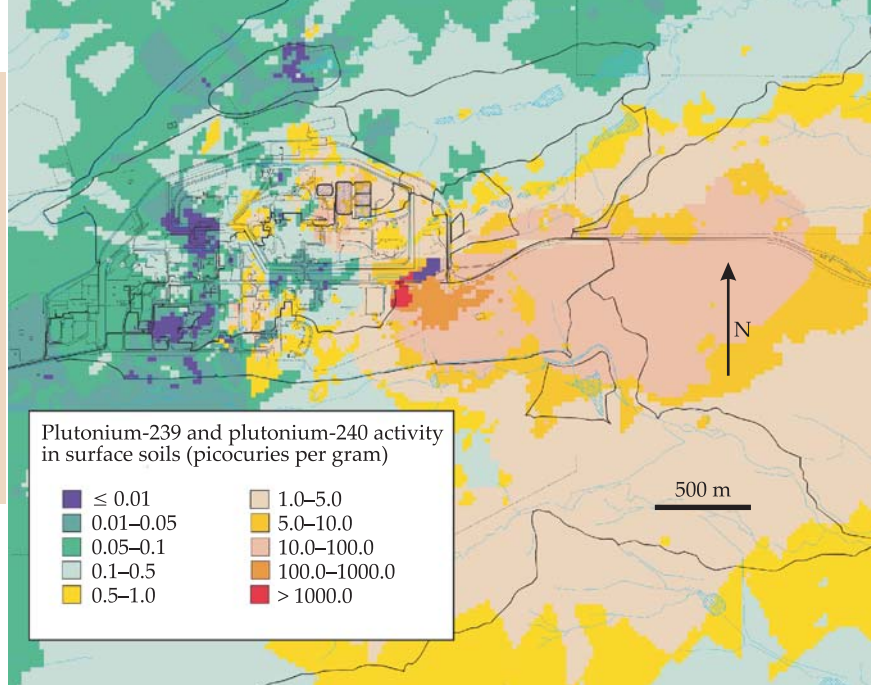
Over its 10-year history, the AME group was led by Christine S. Dayton (Integrated Hydro Systems, previously with Kaiser-Hill), and the following served as advisers: Sumner J. Barr (Los Alamos National Laboratory, retired), Gregory R. Choppin (Florida State University), David L. Clark (LANL), Arokiasamy J. Francis (Brookhaven National Laboratory), Bruce D. Honeyman (Colorado School of Mines), David R. Janecky (LANL), Annie B. Kersting (Lawrence Livermore

National Laboratory), Leonard J. Lane (L. J. Lane Consulting, Inc), D. Kirk Nordstrom (US Geological Survey), and Peter H. Santschi (Texas A&M, Galveston).

The AME group identified and prioritized the technical questions they were to address: What actinide-migration sources and processes are contaminating surface water? What impact would actinide migration have on any planned remedial action? To what standard level do the radioactive sources need to be cleaned? And how will actinide migration affect surface-water quality, air sheds, and downstream areas?

A central principle of AME, even from its inception, was to interact often and openly with the public. As actinide-migration studies got under way, AME representatives met with regulatory agencies and local community and citizens' groups to discuss results and implications for remedial actions at Rocky Flats. Occasionally, additional scientists also took part in discussing technical issues in the public forums, which sometimes prompted further research to address unanswered questions.

Figure 2. To estimate concentrations of plutonium-239 and plutonium-240 in surface soil, researchers applied a geostatistical modeling technique known as Kriging analysis that used nearly 2500 surface-soil samples collected and analyzed between 1991 and 1999.¹⁷ The highest ²³⁹Pu and ²⁴⁰Pu activities—in excess of 1000 picocuries per gram of soil, colored red on the map—were found at the 903 Pad, where plutonium-contaminated solvents had leaked for more than a decade. A clear plume of Pu and Am contamination tracks roughly with the prevailing winds from the northwest to the southeast. Map represents pre-remediation values.



Winds at RFETS predominantly flow from the northwest to the southeast. They can periodically become so strong and gusty—exceeding 160 km/h—that they shatter the windshields of vehicles parked on the site. The wind is an important factor in the dispersal of soil and actinides. Indeed, air monitoring and subsequent calculations of the actinide loads showed that air transport was a dominant actinide migration pathway, before and during cleanup.

Radioactive contaminants

When it was operating, the Rocky Flats nuclear plant generated a huge volume of waste contaminated with radionuclides and other hazardous substances. The majority was shipped offsite, but improper disposal, ruptured or leaking pipes, fires, and faulty storage units resulted in local soil and water contamination. By far the largest source of plutonium and americium contamination in soils emanated from chemical drums stored in an area known as the 903 Pad. Between 1958 and 1969, an estimated 19 000 liters of tainted lathe coolant (about 86 g or 5.3 curies of plutonium) leaked into the ground; wind and surface-water erosion then carried plutonium and americium in a pattern that tracks roughly with the prevailing winds to the east and southeast, at low levels past the eastern site boundary (see figure 2).

Plutonium and americium generally exhibited the same spatial distribution in surface soils, with wide variations in radioactive activities occurring throughout the site. Approximately 90 percent of the radioactive inventory was in the top 12 cm of the soil.³ The concentrations ranged up to several picocuries per liter in streams and ponds, and up to a few nanocuries per gram in soils and sediments.

Chemical reactions, particularly redox reactions in soil and ponds, are often hypothesized to explain actinide mobility. At one extreme, the actinides may react with surrounding materials to create soluble and mobile compounds. At the other extreme, the actinides might remain unchanged at the molecular scale and become bound to natural organic and mineral materials. These natural materials themselves may undergo chemical reactions to form mobile components, thereby carrying the actinides along with them.

The contrast between actinide solubilities—the solubility of plutonium and americium is very low, whereas that of uranium is relatively high—drove researchers' consideration of colloidal and particulate transport processes and prompted the AME advisory group to carefully evaluate evidence that could distinguish solubility and colloidal and particulate results. For example, actinide chemists have long known that under environmental conditions plutonium is

most stable as oxides,⁴ and colloid-sized materials,⁵ but detailed knowledge of their reactivity in the environment is limited to concentrations of picocuries per liter in water and picocuries to nanocuries per gram in soil.

Under natural environmental conditions, plutonium solubility is limited by the formation of amorphous plutonium hydroxide [Pu(OH)₄] or polycrystalline plutonium oxide (PuO₂). Formation of these compounds provides an upper limit on the amount of dissolved—that is, ionic or molecular—plutonium that can be present. Plutonium oxide's measured solubility range⁵ of 10^{–10} to 10^{–13} mol/L is limited by the formation of Pu(OH)₄. Due to that very low solubility and the tendency of compounds of Pu(IV), the fourth oxidation state of plutonium, to adhere to organic and mineral particles, the primary path of plutonium transport is through the migration of fine particles. Indeed, when concentrations of plutonium above fallout levels have been investigated in detail, the plutonium has been linked to colloids and particulates.⁶

Synchrotron radiation studies

Although researchers at Rocky Flats suspected that plutonium contamination in the environment was in a particle form—most likely the very insoluble PuO₂—definitive proof did not exist to verify its chemical form and oxidation state.⁷ Los Alamos National Laboratory researchers led by Steven Conradson performed x-ray absorption fine structure spectroscopy at the Stanford Synchrotron Radiation Laboratory to determine the chemical form of plutonium in RFETS soils and concretes.⁸ Although not well suited for the extremely dilute samples typical of the RFETS environment, the technique successfully identified the chemical fingerprints of select, higher-concentration samples through a careful tuning of the spectroscopy data collection—that is, a judicious choice of which absorption region to analyze, together with long data-collection times.

X-ray absorption near-edge structure analysis identified the oxidation state of plutonium in soils and concretes as Pu(IV). An analysis of the extended x-ray absorption fine structure in the spectra—the spectral oscillations in the region beyond the absorption edge—unambiguously identified the chemical form of plutonium in soil and contaminated concrete around the site as the relatively insoluble hydrous oxide PuO₂·xH₂O (see box 2).

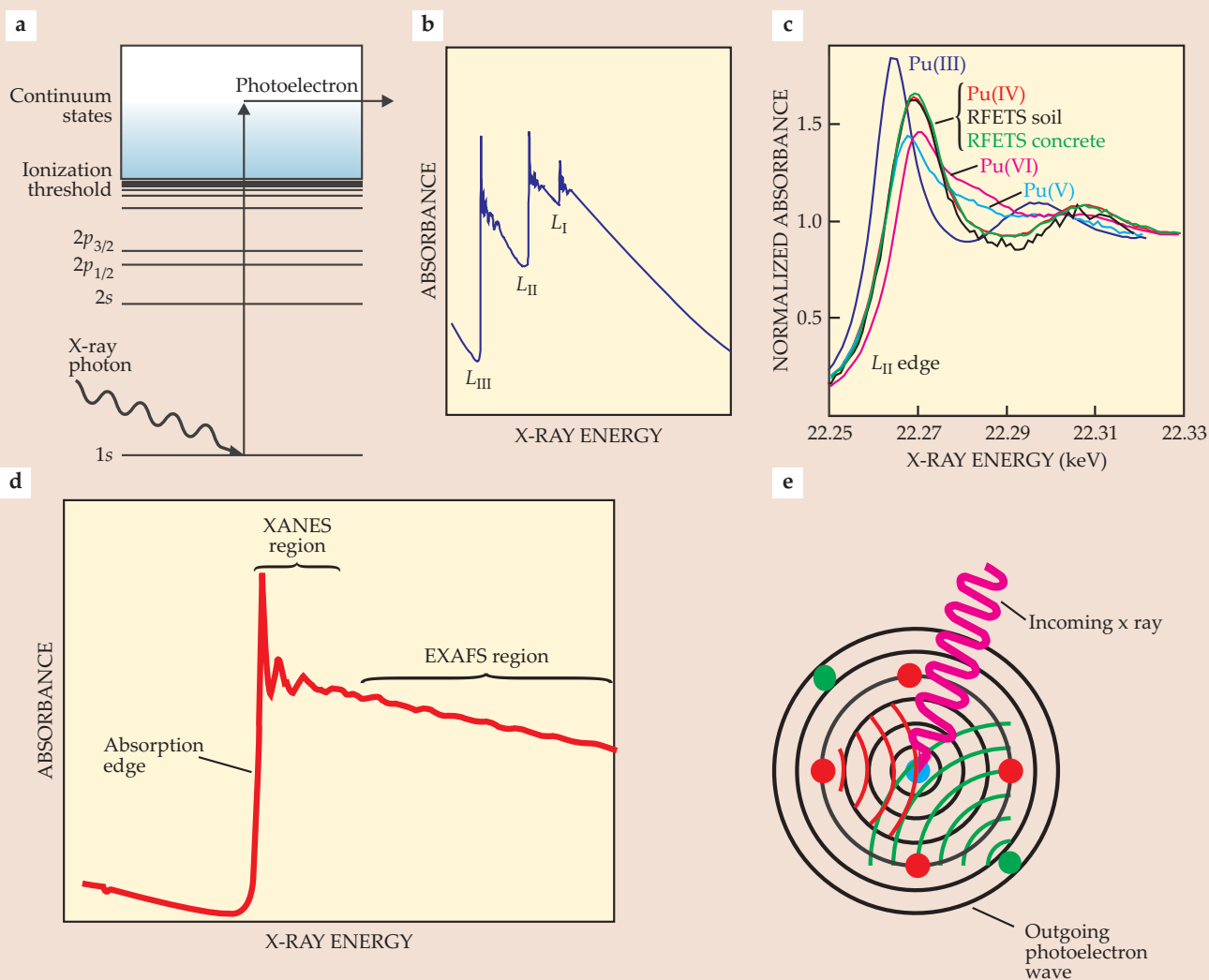
Box 2. X-ray absorption spectroscopy

By monitoring the absorption of x rays in a material as a function of their energy, researchers can map the local atomic structure of the material. Consider the schematic energy-level diagram (a), which pictures an atom's first few core-electron states: $1s$, $2s$, $2p_{1/2}$, $2p_{3/2}$, and so forth. When a core electron absorbs an x-ray photon whose energy is greater than the electron's binding energy, the electron undergoes a transition to an unbound state in the continuum. The abrupt jumps in the simplified absorption spectrum (b) are called absorption edges and correspond to the excitation of an electron from a specific orbital as the x-ray energy becomes sufficient to ionize a core-electron shell. The absorption edge due to excitation of the $1s$ electron is called the K edge; excitations from the less strongly bound $2s$, $2p_{1/2}$, and $2p_{3/2}$ electrons correspond to the L_I , L_{II} , and L_{III} edges, respectively. Although the plutonium L_{III} edge offers the highest absorption intensity, interference from the absorption of other elements from minerals in the samples prompted researchers to focus on plutonium's L_{II} edge, which appears at 22.27 keV.

The near-edge structure, the peaks and shoulders observable over a 20- to 30-eV-wide region just past the edge onset, reveals the oxidation state of the element in molecules and compounds.

An element's ionization potential increases with the ion's valence state, so the absorption generally shifts to higher energy as the oxidation state increases. In the case of Rocky Flats Environmental Technology Site samples—particularly those collected from the 903 Pad soil and concrete exposed to smoke from building fires—the x-ray absorption near-edge structure (XANES) spectra (c) lined up nearly identically with the standard for Pu(IV), the fourth oxidation state of plutonium.

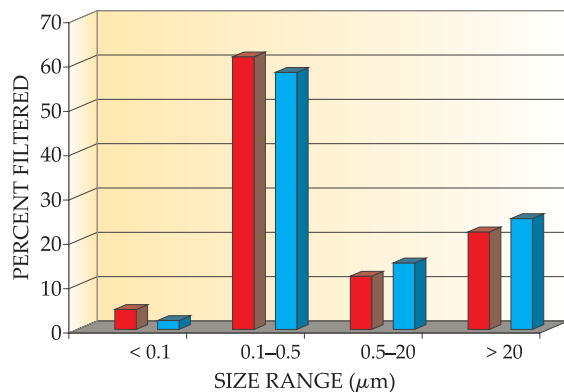
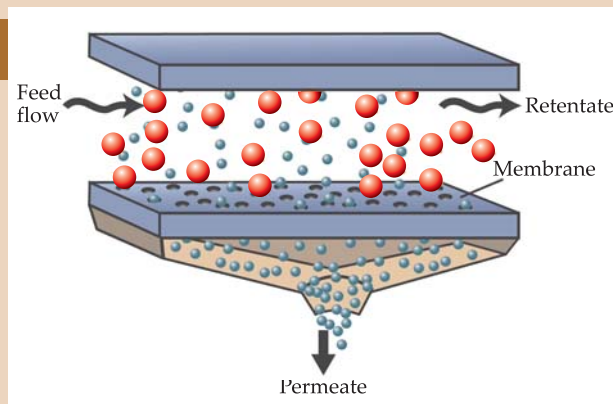
The extended x-ray absorption fine structure (EXAFS), the oscillations that occur beyond the near-edge region, reveals the number of nearest-neighbor bond distances and other structural details of the plutonium-laden samples (see d and e). To appreciate the technique, imagine the case in which a central Pu atom (blue) in PuO_2 absorbs an x-ray photon. The outgoing photoelectron wave (black) scatters off neighboring atoms in the solid (red and green). This backscattered wave then interferes with itself at the original Pu atom. Analyzing the frequency components of the EXAFS oscillations reveals the internuclear bond distances. The EXAFS Fourier transforms of Rocky Flats samples confirmed that the Pu contaminants exist as PuO_2 —in particular, its hydrous form, which includes Pu-OH_2 bonding.



Box 3. Tangential-flow ultrafiltration

In this technique, also called cross-flow ultrafiltration, a particle-filled fluid is pumped along the surface of a membrane, often made of polymer fibers or an inorganic material such as alumina. An applied pressure drives a portion of the fluid—the permeate—through the membrane. The so-called retentate—particulates, colloids, and macromolecules that are too large to pass through—remains on the upstream side. The tangential flow slows the buildup of particles that could clog the pores or reduce the permeate flow by fouling the membrane. Keeping the pores clear is essential for ultrafiltration to reliably separate fine particles. In practice, a broad range of membranes with different pore sizes can filter out, and hence distinguish, the variety of suspended particles, which can range in size from about $1\text{ }\mu\text{m}$ to as small as $0.003\text{ }\mu\text{m}$.

The bar graph shows a summary of ultrafiltration results from Peter Santschi and colleagues¹⁰ that compares normal discharge (blue) at a particular monitoring station with data recorded shortly after a rainstorm (red). The group's measurements demonstrated that a large majority of the plutonium material that passed the $0.5\text{-}\mu\text{m}$ filter was not dissolved, but colloidal, because it could be filtered out using smaller $0.1\text{-}\mu\text{m}$ filters. The experiment showed that suspended matter and colloids came less from eroding soils than from sediment suspended in stream beds.



Judging from the geochemical characteristics found using x rays, one might conclude that insoluble oxides of plutonium and americium would be trapped in the ground and remain immobile. That's true to a point. A growing number of field studies, however, document the movement of low concentrations of low-solubility radionuclides in surface and ground waters.^{6,9,10} Those small concentrations can be transported in surface water and soils by particles of sizes typically ranging from a nanometer to several microns.^{11,12} Because the particles remain suspended in ground water, they can move in the natural watershed and settle into the series of ponds around RFETS.

Ultrafiltration studies

From 1998 until 2001, Texas A&M University's Peter Santschi and coworkers examined ^{239}Pu , ^{240}Pu , and ^{241}Am concentrations in the field and through laboratory studies at RFETS.¹⁰ Since the environmental forms of actinides in the surface waters were in the concentration range of 10^{-3} to 10^{-1} pCi/L, filtration and tangential-flow ultrafiltration were the only methods suitable to separate and analyze the different phases (see box 3). Measurements of total ^{239}Pu , ^{240}Pu , and ^{241}Am concentrations in storm runoff and pond discharge samples collected during spring and summer from 1998 to 2000 demonstrated that most of the ^{239}Pu , ^{240}Pu , and ^{241}Am transported from contaminated soils to streams occurred in the particulate (roughly larger than $0.45\text{ }\mu\text{m}$) and colloidal (roughly between 2 nm to $0.45\text{ }\mu\text{m}$) phases.

In general, most of the Pu and Am in RFETS water was found in the particulate phase, with most of the material that passed a $0.5\text{ }\mu\text{m}$ filter being colloidal. Based on graphite-furnace atomic absorption spectroscopy, transmission electron microscope, and energy dispersive x-ray microprobe

images, colloids were primarily composed of clay and organic matter.

So-called isoelectric focusing experiments of radio-labeled colloids from RFETS soils revealed that colloidal Pu formed in the tetravalent state and was mostly associated with a negatively charged organic colloid having a molecular weight of 10–15 kilodaltons, rather than with the more abundant inorganic colloids made up of iron oxide and clay. Santschi's evidence strongly argued against the presence of mobile colloidal microparticles mainly in the form of PuO_2 , but suggested that PuO_2 is imbedded in, or attached to, organic matter containing some iron.

Each of these complementary studies provided evidence that the low levels of Pu and Am in surface water at RFETS are transported by the colloidal and particulate fraction of the water, not by the dissolved fraction.

Modeling actinide transport

Understanding that Pu and Am exist in the form of insoluble particles clarified that the initial models of contaminant transport—ones based on soluble forms of Pu—were flawed and indefensible. To best fashion the range of possible remediation and management scenarios, AME advisers needed the ability to predict how the radioactive material moved under existing conditions.

AME chose the Water Erosion Prediction Project (WEPP) model,^{13,14} a state-of-the-art process-oriented computer model that simulates hillside erosion processes and estimates the spatial and temporal distributions of soil erosion and sediment deposition in stream channels and impoundments. Because it accounts for enrichment of transported sediment in fine particles, the WEPP model is well suited for contaminant transport calculations.

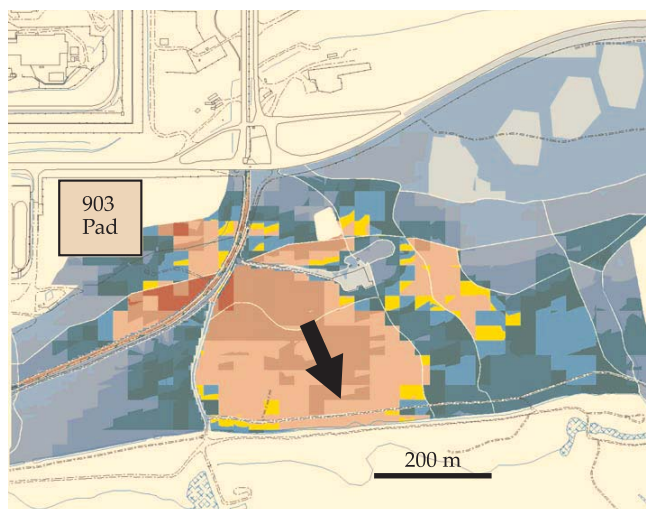


Figure 3. Plutonium mobility map of a severe 6-hour storm event, modeled for the 903 Pad and its local watershed. Red indicates the highest plutonium mobility, blue the lowest. The models indicate that up to 99% of Pu in the surface water that flows into an interceptor ditch (below the colored region of the figure) comes from hillside erosion. After the water, sediment, and Pu are delivered to the ditch's stream channel, a sediment-transport model was used to predict the route and deposition of plutonium-laden sediment downstream. The arrow points in the direction of sediment transport.

To estimate stream channel sediment erosion and deposition, output from the WEPP model was routed into yet another—the US Army's Hydrologic Engineering Center sediment-transport model, HEC-6T, which can accommodate up to 100 tributaries flowing into a main channel.¹⁵ The combination was crucial to modeling the RFETS watersheds and using soil data to predict surface-water actinide concentrations.

AME applied the soil-erosion and sediment-transport models to the hillslopes and channel systems at RFETS and compared the results with monitoring data to parameterize, initialize, and calibrate the models. The coupled models could then be used to simulate storm events and the transport of ²³⁹Pu, ²⁴⁰Pu, and ²⁴¹Am contaminants, estimate the amount of contaminated sediment in surface water, and analyze which hillslopes and drainages the contamination moved along. Finally, the coupled models were used with data on climate and soil contamination to predict rates of sediment and contaminant transport under various management scenarios designed to handle the cleanup.

As part of the modeling process, the predicted soil erosion—that is, the mass eroded per unit area—was combined with actinide soil-concentration data to generate a map of actinide mobility predicted for a specific storm event. Surprisingly, the results of those maps revealed that the largest Pu and Am loads delivered to surface water do not necessarily originate from areas with the highest concentrations of Pu and Am in the soil. The combination of topography, vegetative cover protecting the soil, soil erodibility, and actinide concentration determines the rate of erosion and contaminant transport.

The area east of the 903 Pad, for example, generally contains the highest levels of Pu and Am in the area (see figure 3). The area around the 903 Pad, however, is relatively flat, with slopes of only about 1%. Consequently, that area suffers far less soil erosion by water than other, steeper parts of the watershed—with a corresponding reduction in the amount of Pu and Am transported.

Cleanup

The scientific understanding developed through the integrated studies described above clarified the issues surrounding Pu and Am migration in the RFETS environment. Once Kaiser-Hill, DOE, the EPA, the Colorado state and local governments, and concerned citizens' groups reached a common appreciation of the technical issues, the different groups could then reach long-sought agreements on how to proceed with cleanup. Realizing that Pu and Am existed primarily in particulate forms led to an understanding of their movement

at the site via wind and water. That set the stage for discussing the potential risks to human health and the environment, possible remediation efforts, specific soil-removal technologies, and ways to best reconfigure the landscape.

Site operators responded with a major shift of emphasis to soil erosion and the need to control it. The most poignant illustration of that shift was a management directive distributed to every employee from Kaiser-Hill president Nancy Tuor; the directive discussed preventing the dispersal of contaminants during remediation efforts and reducing the transport of Pu and Am to nearby stream channels or locations off site. Such measures allowed site remediation to proceed rapidly and thus meet or beat deadlines.

In 1996 the maximum allowable radionuclide action level was 651 pCi/g. In 2002, armed with improved understanding of Pu behavior, DOE, the Colorado Department of Public Health, and the EPA released a series of reports that formed the basis for a new maximum surface-soil action level of 50 pCi/g; that standard was based on risk analysis and was the result of huge community involvement. Because the Pu contamination was generally confined to surface soils, the greatest public health risk came from the forces of wind and water. In actual decontamination, demolition, and remediation, workers therefore set up large tents at the 903 Pad to insulate work in progress from wind, rainfall, and erosion. The work focused on removing soil contaminated at the more aggressive standard, down to one meter below the surface, and replacing it with fresh soil; soil contaminated at depths greater than one meter was allowed to remain in place, even at higher concentrations. To decontaminate the concrete walls of buildings, workers used a variety of techniques, including pressure washing of the top layers to remove the radioactive particles. They then used the clean concrete as backfill around the site.

Operators developed a storm-water pollution-prevention plan, designed to minimize the erosion, sedimentation, and runoff of water across the site. Erosion-control measures included straw bales and wattles, straw crimping, silt fences, mats, hydromulch and crimped synthetic fibers (Flexterra), and riprap lining of drainage channels. Some new wetland areas were also prepared.

As a result of the cleanup activities and control measures, surface water and air monitoring stations at the site boundary have actually shown a decrease in actinide migration. Several of those measures are expected to work only for a few months to a few years, and will require regular maintenance until the region stabilizes and the vegetation is reestablished.

A new paradigm?

Superfund sites, such as RFETS, represent important environmental problems of national significance. So it is important that our best science is applied to improve the technical basis for decision making.¹⁶ A confluence of several fortunate

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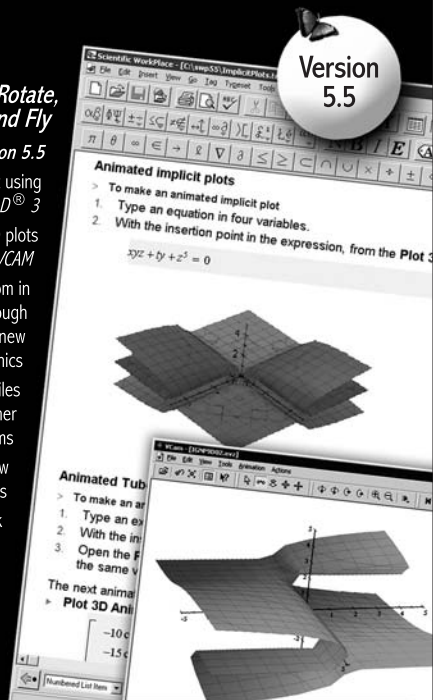
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factors made the RFETS cleanup successful: the willingness of Kaiser-Hill to seek outside scientific advice; the acceptance, down to the project level, of the value of that advice in avoiding pitfalls and improving operations; and stakeholders' acceptance, albeit more gradual, of the independence and veracity of the AME scientific advisers. This willingness and acceptance helped DOE, the integrating contractor, regulators, and the involved community to focus on specified goals and objectives.

Establishing particle-transport mechanisms as the basis of Pu and Am mobility, rather than aqueous sorption-desorption processes, provided a successful scientific foundation for understanding the scope and nature of the problem and how best to solve it using erosion control technology. The understanding prompted contractors to rapidly apply soil-erosion and sediment-transport models. That, in turn, led to the design and sitewide use of erosion control technology to mitigate the transport of radioactive particles. Moreover, a scientific understanding of the problems helped define a clearer endpoint and led to the most extensive cleanup in the history of Superfund legislation. Consequently, the project finished one year ahead of schedule, saved taxpayers billions of dollars, and removed an annual liability of more than \$600 million from the DOE budget.

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